

2017-08-16

Aeolian sediment fingerprinting using a Bayesian mixing model

Gholami, H

<http://hdl.handle.net/10026.1/9893>

10.1002/esp.4189

Earth Surface Processes and Landforms

Wiley

All content in PEARL is protected by copyright law. Author manuscripts are made available in accordance with publisher policies. Please cite only the published version using the details provided on the item record or document. In the absence of an open licence (e.g. Creative Commons), permissions for further reuse of content should be sought from the publisher or author.

**Earth Surface
Processes and Landforms**

**Aeolian sediment fingerprinting using a Bayesian mixing
model**

Journal:	<i>Earth Surface Processes and Landforms</i>
Manuscript ID	ESP-17-0002.R1
Wiley - Manuscript type:	Research Article
Date Submitted by the Author:	n/a
Complete List of Authors:	Gholami, Hamid; University of Hormozgan, Department of Range and Watershed Management Telfer, Matt; Plymouth University, School of Geography, Earth and Environmental Sciences Blake, William; Plymouth University, SoGEES Fathabadi, Abolhassan; University of Gonbad-e-Kavoos, Department of Range and Watershed Management
Keywords:	Aeolian sediment, Sand provenance, Markov Chain Monte Carlo, Fingerprinting, Dune

SCHOLARONE™
Manuscripts

Aeolian sediment fingerprinting using a Bayesian mixing model

Hamid Gholami ^{a*}, Matt W. Telfer ^{b*}, William H. Blake ^b and Abolhassan Fathabadi ^c

^a Department of Range and Watershed Management, University of Hormozgan, Bandar-Abbas, Hormozgan, Iran.

^b School of Geography, Earth and Environmental Sciences, Plymouth University, Plymouth, Devon, PL4 8AA , UK.

^c Department of Range and Watershed Management, University of Gonbad-e-Kavoos, Gonbad-e-Kavoos, Golestan, Iran.

* Correspondence to:

Hamid Gholami, Department of Range and Watershed Management, University of Hormozgan, Bandar-Abbas, Hormozgan, Iran. *E-mail address:* hgholami@hormozgan.ac.ir. Tel: +98 937 0865077.

and

Matt Telfer, School of Geography, Earth and Environmental Science, Plymouth University, Plymouth, Devon, PL4 8AA, UK. *E-mail address:* matt.telfer@plymouth.ac.uk. Tel: +44 1752 585570.

Abstract

Identifying sand provenance in depositional aeolian environments (e.g. dunefields) can elucidate sediment pathways and fluxes, and inform potential land management strategies where windblown sand and dust is a hazard to health and infrastructure.

1
2
3 23 However, the complexity of these pathways typically makes this a challenging
4
5 24 proposition, and uncertainties on the composition of mixed-source sediments are
6
7 25 often not reported. This study demonstrates that a quantitative fingerprinting method
8
9
10 26 within the Bayesian Markov Chain Monte Carlo (MCMC) framework offers great
11
12 27 potential for exploring the provenance and uncertainties associated with aeolian
13
14 28 sands.

15
16 29 Eight samples were taken from dunes of the small (~58 km²) Ashkzar erg, central
17
18 30 Iran, and forty-nine from three distinct potential sediment sources from the
19
20 31 surrounding area. These were analyzed for 61 tracers including 53 geochemical
21
22 32 elements (trace, major and rare earth elements (REE)) and 8 REE ratios. Kruskal–
23
24 33 Wallis H-tests and stepwise discriminant function analysis (DFA) allowed the
25
26 34 identification of an optimum composite fingerprint based on six tracers (Rb, Sr, ⁸⁷Sr,
27
28 35 (La/Yb)_n, Ga and δCe), and a Bayesian mixing model was applied to derive the
29
30 36 source apportionment estimates within an uncertainty framework.

31
32
33 37 There is substantial variation in the uncertainties in the fingerprinting results, with
34
35 38 some samples yielding clear discrimination of components, and some with less clear
36
37 39 fingerprints. Quaternary terraces and fans contribute the largest component to the
38
39 40 dunes, but they are also the most extensive surrounding unit; clay flats and marls,
40
41 41 however, contribute out of proportion to their small outcrop extent. The successful
42
43 42 application of these methods to aeolian sediment deposits demonstrates their
44
45 43 potential for providing quantitative estimates of aeolian sediment provenances in
46
47 44 other mixed-source arid settings, and may prove especially beneficial where
48
49 45 sediment is derived from multiple sources, or where other methods of provenance
50
51 46 (e.g. detrital zircon U-Pb dating) are not possible due to mineralogical constraints.
52
53
54
55
56
57
58
59
60

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

47 **Key words:** Sand provenance; Aeolian sediment; Markov Chain Monte Carlo;
48 Fingerprinting; uncertainty.

For Peer Review

1. Introduction

Identifying and quantifying the source(s) of aeolian sediments is a long-standing challenge for geoscientists, and yet such information is often of crucial importance in understanding sediment fluxes at a range of scales. As well as providing fundamental knowledge on long-term landscape evolution (e.g. Pell et al., 1997), aeolian provenance studies have been used to elucidate past wind regimes and palaeoclimates (e.g. Nanson et al., 1995), investigate hazardous dust transport pathways (e.g. Pethierick et al., 2008; Yang et al., 2007) and inform studies of the palaeoclimatic record of the Antarctic ice-core dust record (Delmonte et al., 2010; Delmonte et al., 2004). The challenges with the task arise not just from the diverse range of potential sources for aeolian sands and dusts (e.g. geological or lithological units, soil units, land use types and geomorphological landscapes), and the long transport distances which may be involved (on the order of $10\text{-}10^2$ km for aeolian sand and $10\text{-}10^3$ km for aeolian dust), but also the potential complexity of transport pathways (Huntsman-Mapila et al., 2005). Before deposition at its current location, an aeolian sand grain may have been through multiple cycles of fluvial, aeolian, lacustrine and/or colluvial deposition and subsequent mobilization. Aeolian sands, therefore, rarely retain an easy-to-interpret signature of their origins.

The use of geochemical fingerprinting methods to determine sediment provenance has progressively increased since the late 1990s (Walling, 2013). Application has been focused most widely in fluvial contexts (Haddadchi et al 2013) wherein there recent work has highlighted the need to pay attention to challenges in signature development and tracer behavior (Koiter et al., 2013). Sediment fingerprinting involves the identification, quantification and statistical testing of a range of source

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

74 material properties capable of discriminating between potential sediment sources
75 with a view to improving knowledge of sediment source and transport processes
76 (Collins et al., 2017) . These properties may include geochemical characteristics
77 (e.g. Douglas et al., 2009; Lin et al., 2015), radionuclide concentrations (Wilson et
78 al., 2012), mineralogy (Pittam et al., 2009), geochronological data (principally U-Pb
79 dating of detrital zircons; e.g. Pell et al., 1997; Garzanti et al., 2013), biomarkers
80 (Chen et al., 2016) and colour properties (Martínez-carreras et al., 2010). Although
81 sediment fingerprinting studies of aeolian sands are not new (e.g. Pell et al., 1997;
82 Wasklewicz and Meek, 1995; Winspear and Pye, 1996; Liu et al., 2016; Muhs et al,
83 2017), challenges remain in adequately capturing the uncertainties associated with
84 the diverse sources and pathways that may exist, and adoption of techniques
85 developed in different disciplines offer a route forward. It is noteworthy that in a
86 recent review discussing applications of sediment source-tracing methods (Owens et
87 al., 2016), mention of aeolian sedimentation is limited to health science studies of
88 PM2.5 and PM10 material. The opportunity to utilize these approaches in aeolian
89 process science remains largely overlooked.
90
91 In recent years, increasing attention has been directed to the uncertainty of the
92 results generated by sediment source fingerprinting. It is important that such
93 uncertainty is recognized, particularly if the results are to be used to target
94 investment in sediment control measures (Mukundan et al., 2012). The factors
95 contributing to uncertainty in estimates of source apportionment are manifold and
96 diverse, and have been reviewed elsewhere (e.g. Walling 2010; Koiter et al., 2013;
97 Collins et al, 2017); here we consider some of the differences between the fluvial
98 setting of most sediment fingerprinting studies, and the aeolian context considered

99 here. Many uncertainties remain the same – for instance, instrumental precision.
100 Other aspects of the aeolian entrainment, transport and depositional system,
101 however, differ markedly from fluvial settings. Due to dominance of gravity in
102 controlling - and directing - slope and fluvial processes, and the (usually) confined
103 nature of fluvial systems, erosion and entrainment of sediment from catchments is
104 highly directionally-controlled. In an aeolian context, there is more scope for spatially
105 extensive direct entrainment of sediment, and also more potential for directional
106 variability, and this complex mixing environment may in turn lead to increased
107 covariance between properties used to derive the fingerprint. In addition, such
108 complexities cannot be considered static, as variations in wind regimes over long
109 timescales may lead to changes in these pathways.

110
111 In order to quantify the uncertainty associated to mixing models related to this
112 inherent variability in the source area and sediment mixture data, some recent
113 studies have explored the use of Monte Carlo simulations (e.g. Motha et al., 2003;
114 Collins et al., 2013; Collins et al., 2012; Stone et al., 2014; Smith & Blake, 2014;
115 Sherriff et al., 2015; Vale et al., 2016; Gellis & Noe, 2013; Voli et al., 2013; Wilkinson
116 et al., 2013; Walling et al., 2008). More recently, Bayesian mixing models being
117 employed more comprehensively translate component uncertainties into source
118 apportionment results (Cooper and Krueger, 2017) with several examples
119 undertaken in hydrological contexts (e.g. Fox & Papanicolaou, 2008; Cooper et
120 al., 2014; Cooper et al., 2015; Nosrati et al., 2014; Stewart et al., 2015). To date,
121 however, such approaches have not been used within aeolian sedimentary contexts.

122

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

The sophistication of aeolian sediment provenance studies currently lags those in the fluvial sphere, and the main aim of this paper is to demonstrate the viability of fingerprinting methods for aeolian sediments, including the estimation of uncertainty, associated with the contributions from different geological units as potential sources for a small dunefield, Ashkzar Erg, in the Yazd-Ardekan Plain, central Iran, using a Bayesian mixing model. Ashkzar erg and its surrounding potential sources cause many serious problems related to wind erosion and associated on-site and off-site effects, with potential impacts on the health of the occupants of the neighbouring city of Yazd (Naddafi et al., 2006). Aeolian deflation is a major erosional process on the Yazd-Ardekan Plain and large amounts of aeolian sediment are often transported to residential regions by wind (Amiraslani and Dragovich, 2011). Therefore, quantifying sediment source contributions to the Ashkzar sand dunes could help to select the best management strategies at this location and others similarly affected by aeolian erosion. Additionally, the findings are considered in their geomorphological context with the aim of explaining the spatial variability in sediment provenance observed at Ashkzar erg.

2. Materials and methods

2.1. Field location

Yazd-Ardekan (31°10'–32°43' N, 53°68–54°47' E) is an arid plain in central Iran, and includes different geomorphic landscapes, such as the Ashkzar and Yazd ergs (Figure 1). The Yazd-Ardekan Plain is surrounded by mountain ranges. These are Shirkooch in the south, Ahangaran and the Margh Zard Mountains in the west, Haft Adamin and Khoonzad Mountains in the east and Chak Chak Mountain in the north. The area of the plain is ~2900 km², and it consists of 78% Quaternary alluvial fans

1
2
3 148 and terraces (Qt₂ geological unit), 13% clay flats (Qc geological unit), 7% Eocene
4
5 149 gypsiferous marl (Egm geological unit) and 2% sand dunes (Qsd geological unit)
6
7 150 (Figure 1 and 2). About 93% of Yazd-Ardekan Plain is thus covered by Quaternary
8
9 151 deposits. Active and stabilized sand dunes in the Ashkzar erg occupy 58 km²
10
11 152 (centred on 32° 1'N, 54° 10'E), which is dominated by barchans and transverse
12
13 153 barchanoid ridges (Figure 2). The erg has a sparse but extensive cover of *Haloxylon*
14
15 154 *persicum*, a species which is both endemic to the region, and is also used to stabilize
16
17 155 mobile sand (Amiraslani and Dragovich, 2011). Based on 50 years of climate data at
18
19 156 Yazd Meteorological Station, minimum and maximum annual temperatures are -16
20
21 157 °C and 46 °C, respectively. Long-term mean rainfall and annual evaporation in the
22
23 158 study area are ~60 mm/year and ~3500 mm/year respectively. According to annual
24
25 159 wind roses, dominant winds on the Yazd-Ardekan Plain are mainly from the north
26
27 160 and west (Figure 1C).
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

161
162 [Approx. location of Figure 1]
163

164 2.2. Sampling and laboratory analysis

165 The geological units that were identified as potential sources for sand dunes (Qsd
166 formation) are the Qt₂ (Quaternary alluvial fans and terraces), Qc (Quaternary clay
167 pans and flats) and Egm (Eocene marls) formations. Other surrounding lithologies in
168 the vicinity are hard, igneous exposures and can be discounted from generating
169 substantial quantities of deflatable sediment. In this study, spatially distributed
170 source samples were taken from 49 sites, covering the Egm (n=8), Qc (n=18) and
171 Qt₂ (n=23) potential sources, and eight sediment samples were collected from the
172 Ashkzar sand dunes (Figure 1D). Samples were collected from the upper 0–5 cm

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

173 depth of potential sources (that is, the layer of the regolith exposed to current aeolian
174 entrainment) and sand dunes (that is, the layer of the regolith most recently
175 deposited); this is similar to sampling strategies employed by other provenance
176 studies of aeolian dunes (e.g. Pell et al., 1997), and is accordance with common
177 earth science protocols (Owens et al., 2016). Within each source area, sample
178 selection was based upon locations that were: a) clearly derived from the geological
179 unit in question, b) selected to ensure broad spatial coverage of the source area, and
180 c) clearly influenced by aeolian erosion (e.g. the presence of deflatable unvegetated
181 sand surfaces with ripples, as well as yardangs of a range of scales). Samples
182 numbers were chosen to ensure a balance between the greater spatial extent of the
183 Qt2 unit (i.e. sampling was stratified), whilst maintaining a minimum of eight samples
184 for the smaller Egm and Qc units. The spatial location of sampling sites is shown in
185 Figure 1D.

186 All sand dune and potential source samples were dry sieved for particle size data,
187 and to isolate the 62.5-150 μm fraction for further geochemical analysis. This fraction
188 was chosen as it represents the dominant fraction in each of the dune samples
189 (Table 1), and is of a size range susceptible to aeolian transport (whilst excluding
190 any contribution of larger grains from local sources, either by aeolian creep or other
191 transport processes). Concentrations of elements including major, trace and rare
192 earth elements (REE) were determined using ICP-MS, after direct digestion with
193 aqua regia (e.g. Collins et al., 2010; Collins et al., 2012); and concentration of
194 strontium and neodymium isotopes measured by ICP-MS, after digestion with a
195 mixture of $\text{HNO}_3 + \text{HClO}_4 + \text{HF}$ (3:2:1) (e.g. Honda et al., 2004; Rao et al., 2011). The
196 relative standard deviation (%RSD), based on three replicates for each determinant
197 on each sample, was consistently $\leq 4\%$. With regards to REE concentrations, eight

1
2
3 198 REE ratios including Σ REE, Nd/Yb, Eu/Eu* (Europium Anomaly), (La/Lu)_n, (La/Sm)_n,
4
5 199 (Gd/Yb)_n, (La/Yb)_n and δ Ce (Cerium Anomaly) were calculated (e.g. Daga et al.,
6
7 200 2008; Dou et al., 2010; Rao et al., 2011). In total, 61 tracers were used to fingerprint
8
9 201 the sediments of the Yazd-Ardekan Plain.
10
11
12 202

13
14 203 [Approx. location of Table 1]
15

16 204 [Approx. location of Figure 2]
17

18 205 [Approx. location of Figure 3]
19

20
21 206

22 207 *2.3. Discrimination of aeolian sediment sources*

23
24
25 208 We employed a two-stage statistical method proposed by Collins and Walling (2007)
26
27 209 to characterize the composite fingerprint for the sources of the sands of the Ashkzar
28
29 210 dunes. In stage one, all individual fingerprint properties were tested for their ability to
30
31 211 distinguish source types, using the Kruskal–Wallis H-test. Properties with critical
32
33 212 values at the 95% level of confidence could be used in a composite fingerprinting
34
35 213 model to discriminate between sources types. In stage two, stepwise discriminant
36
37 214 function analysis (DFA) was employed to identify the optimum composite fingerprint
38
39 215 model from the properties selected in stage one. The stepwise DFA was based on
40
41 216 the minimization of Wilk's lambda was used to select optimum composite fingerprint.
42
43 217 The F values were used as the test criteria to enter and remove elements. The
44
45 218 threshold of F value for entering and removing of elements was set to 3.84 and 2.71,
46
47 219 respectively (e.g. Vale et al, 2016).
48
49
50
51
52

53 220 *2.4. Bayesian mixing model*

54
55
56
57
58
59
60

End-member mixing models have been taken a variety of approaches to account for uncertainty in the mixing model (Cooper and Krueger, 2017) and some (e.g. Brewer et al., 2005; Fox & Papanicolaou, 2008) have adopted hierarchical Bayesian models, which we adopt here. Within the mixing model formulation, we assume that, for each source s , the sample i tracer composition, x , has a multivariate normal distribution as follows:

$$x_s^i \sim \text{MVN}_A(\mu_s, \Sigma_s), \quad s = 1, \dots, N, \quad i = 1, \dots, n_{x,s} \quad (\text{eq. 1})$$

where $n_{x,s}$ indicates the number of samples of source s ; μ_s is a A -dimensional vector representing mean fingerprints for source s ; Σ_s represents a $(A \times A)$ dimensional covariance matrix for source s . There are n_z sediment samples for which A fingerprints were measured for each sample $Z^j = (z_1^j, \dots, z_A^j)^T, j = 1, \dots, n_z$ and these fingerprints have multivariate normal distributions:

$$Z^j \sim \text{MVN}_A(\mu_j^z, \Sigma^z) \quad (\text{eq. 2})$$

Each source s has a fractional contribution p_s^j to each sediment sample j . The contribution of source types for each sediment sample is equal to $p_s^j y_s^j$, where y_s^j is an unobserved (latent) variable that follow the same distribution as X_s^i .

$$\mu_j^z = \sum_{s=1}^N p_s^j y_s^j, \quad j = 1, \dots, n_z \quad (\text{eq. 3})$$

$$\sum_{s=1}^N p_s^j = 1, \quad 0 \leq p_s^j \leq 1 \quad (\text{eq. 4})$$

Each fractional contribution must be between zero and one, positive and all of them must sum to unity. To meet this constraint, some studies have used Dirichlet distribution as a prior for the fractional contribution (e.g. Fox & Papanicolaou, 2008; Massoudiehet al., 2013), whereas other studies used transformation such as

centered log-ratio (CLR) (Senn et al., 2009), isometric log-ratio (ILR) (e.g. Cooper et al., 2015; Parnell et al., 2013; Egozcue et al., 2003) and additive log-ratio (ALR) (e.g. Brewer et al., 2005; Palmer & Douglas, 2008). In this study, a CLR transformation was used, as it has been shown to produce comparable median values to other methods, but with better precision (Cooper et al., 2014). The transformation applied is thus:

$$\phi_i = CLR(P_i) = \log\left[\frac{P_{i1}}{g(P_i)}, \dots, \frac{P_{ik}}{g(P_i)}\right] \quad (\text{eq. 5})$$

$$\phi_i \sim (\mu_\phi, \tau_\phi) \quad (\text{eq. 6})$$

where $g(p_i)$ is the geometric mean of the proportion vector. Figure 4 shows a directed acyclic graph of the model. Compared to an empirical Bayesian approach in which some prior parameters are estimated using deterministic data, the full Bayesian approach employed here needs to specify prior distribution for all parameters. When there is little information about the parameters, using informative hyper-parameters cause biased results. In this study, weakly or non-informative hyper-parameters were used. Multivariate normal and inverse-Wishart distributions were selected as prior distribution for sources means and covariance matrix, respectively.

$$\mu_s^X \sim MVN(\theta_s, \tau_s^{-1}), \quad s = 1, \dots, N \quad (\text{eq.7})$$

$$\Sigma_s^X \sim \text{Inverse} - \text{Wishart}(\Omega_s^X, \rho_s^X), \quad s = 1, \dots, N \quad (\text{eq.8})$$

Here, the hyper-parameter θ_s was set to the sample means of the fingerprints and τ_s was set as a diagonal matrix with values 0.01 on the diagonal. For Wishart distribution, the hyper-parameter Ω_s^X is a diagonal matrix with value 1 as diagonal

elements and ρ_S^X was set to six (to reflect the lack of information on the precision matrices, and the number of tracers selected for the fingerprint). Similar prior distribution and hyper-parameters was assigned for sediment covariance matrix.

$$\Sigma^Z \sim \text{Inverse} - \text{Wishart}(\Omega, \rho) \quad (\text{eq.9})$$

Weakly informative hyper-parameters $N(0,1)$ and $\text{Inv-}\Gamma(2,1)$ were assigned for μ_ϕ and τ_ϕ , respectively.

The complete posterior distribution of all model parameters for sediment sample Z_j can thus be written as

$$\begin{aligned} (\Sigma_S, \tau_\phi, \mu_\phi, \Sigma^Z, p_j, \phi, \mu_S | X, Z_j) &\propto \prod_{S=1}^N \prod_{i=1}^{n_{x,s}} \{P(x_s^i | \mu_S, \Sigma_S)\} \times \prod_{S=1}^N P(\mu_S | \theta_S, \tau_S^{-1}) \times \\ &\prod_{S=1}^N P(\Sigma_S | \Omega_S, \rho_S) \times P(Z_j | \mu_j^Z, \Sigma^Z) \times P(\Sigma^Z | \Omega, \rho) \times P(\phi | \mu_\phi, \tau_\phi) \times P(\mu_\phi) \times P(\tau_\phi) \end{aligned} \quad (\text{eq.10})$$

As the joint posterior of all parameters is complex and high-dimensional, we cannot directly obtain posterior distribution functions, but the Bayesian model thus defined can be analyzed using Markov Chain Monte Carlo (MCMC); we have used the WinBUGS package (Lunn et al., 2000) to derive parameter estimates. MCMC methods require that the chain reaches a steady state, and the number of runs required to reach this state is considered as burn in. The model was run by taking 50,000,000 times from the posterior distribution from the sand dune and source samples, and the first 5,000,000 runs were considered as burn in. The large number of iterations was used to ensure convergence, despite the model's complexity and high dimensionality; the model converged during the run, as assessed by trace plots of simulations, Monte Carlo error and autocorrelation.

[Approx. location of Figure 4]

289

3. Results

Grain size data are presented in Table 1 to enable consideration of potential sorting effects during aeolian transportation. The sources reveal very similar physical grain sizes, and it is worth noting that whilst the Qc unit is mapped as a 'clay flat', the sediment sampled for analysis is dominantly sand. The erg, on the other hand, as might be expected as a result of aeolian transport and deposition is better sorted, and less enriched in the coarse and very coarse sand fraction. The individual dune sand samples retain marked variability, with the very-fine (62.5-150 μm) fraction ranging from 37% to 65%, and a single sample (8) retaining a substantial coarse (> 600 μm) component.

300

The Kruskal-Wallis H test (i.e. one-way ANOVA) was performed on geological units Egm, Qc and Qt2. Results identified 25 significant tracers between these groups (Table 2). Tracers that failed this test ($p > 0.05$) were removed. These were: Nd, Sm, Gd, Tb, Dy, Yb, Lu, (Nd/Yb), (Gd/Yb)_n, (La/Sm)_n, V, Cr, Co, Ni, Cu, Zn, Y, Zr, Nb, Ta, U, As, Bi, Cd, Ge, In, Mo, Sb, Se, Te, W, Mn, Si, ¹⁴³Nd, ¹⁴⁴Nd and ⁸⁶Sr. Whilst the successful discrimination of different tracers between geological units will vary when this method is applied to settings other than this location, the presence of 25 tracers with significant discriminatory power suggests that this method may be applicable in diverse geological settings and/or areas with contrasting weathering regimes.

310

[Approx. location of Table 2]

312

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

313 According to the DFA, a total of six individual tracer properties (Rb, Sr, ⁸⁷Sr,
314 (La/Yb)_n, Ga and δCe) were selected for the optimum composite fingerprint, which
315 correctly discriminated 81.6% of the source type samples (Figure 5).

316

317 [Approx. location of Table 3]

318 [Approx. location of Figure 5]

319

320 Although DFA results suggested that good source discrimination was achieved, with
321 clear separation of the three group centroids, samples sourced from Qc were found
322 to slightly overlap with the Qt2 source when the first two discriminant functions were
323 plotted, and, to a lesser degree Qt2 and Egm also overlap slightly (Figure 5). The
324 mean and SD of six optimum composite fingerprints that were selected for the
325 Bayesian mixing model, are presented in Table 3. These were tested for normality
326 via Wilks-Shapiro tests (Table 4), and the raw data revealed that the Sr and δCe
327 tracers did not follow a normal distribution for all settings. To account for this, Box-
328 Cox transformations (Box and Cox, 1964) were applied to all data, and the
329 transformed data were used for model experimentation.

330

331 [Approx. location of Table 4]

332

333 The derived source contributions for the eight sand dune samples are presented in
334 Table 5 and Figure 6. Overall, the alluvial fans and terraces (Qt2) provide the most
335 abundant supply of sands (mean contribution across all 8 samples = 45.4%, and
336 locally up to 92.7%), with the clay pans (Qc) and Eocene marls (Egm) each
337 contributing around a quarter of the net sediment aeolian flux. However, the
338 composition of the dune sands is highly variable, with different samples dominated

339 by different contributing sources, and locally, all three of the potential sources occur
340 as both maxima and minima.

341

342 [Approx. location of Figure 6]

343

344 4. Discussion

345 4.1 Development of a Bayesian mixing model to discriminate aeolian sediment 346 pathways

347 The mixing model to fingerprint aeolian sediment sources deployed in this study
348 used composite signature comprising a suite of six geochemical characteristics (Rb,
349 Sr, ^{87}Sr , La:Yb, Ga and δCe) identified by stepwise DFA as the most appropriate,
350 and was able to account for >82% of the variance between the three sources. The
351 suite of properties selected by the DFA method most likely reflects two principal
352 factors; the ultimate source of the sediments, and the degree of weathering. The
353 latter might well have variable influence across the source areas, and hence there is
354 some overlap between samples of each class. High La:Yb ratios, for instance, are
355 typically associated with deep igneous lithogenesis (Deffant and Drummond, 1990)
356 and may locally reflect differing sediment contributions from the Precambrian
357 crystalline basement provinces of central Iran. δCe , similarly, is often associated with
358 intrusive igneous rocks, although may also be enriched in some sedimentary rocks
359 (Wedepohl, 1978); in this study, the highest concentrations are found in the alluvial
360 fans derived from the adjacent igneous mountains, but the second highest
361 concentrations are found in the sedimentary marls of the Egm unit (Figure 2). In
362 short, the highly varied geology of central Iran, ranging from Precambrian magmatic

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

363 rocks to Cenozoic marine sediments, promotes a high degree of variance in the
364 geochemical fingerprint of modern aeolian sediments. Compounding this is the range
365 of weathering intensities seen, from the intense weathering history of the sediments
366 of Quaternary clay pans, to the more moderate weathering of the sands of the
367 alluvial fans forming the piedmonts of the neighbouring ranges.

368 4.2 Potential for application to other aeolian depositional settings

369 Despite the usefulness of understanding the provenance of aeolian sands, the
370 sophistication of unmixing models within the aeolian science community generally
371 lags that of fluvial science, in particular in terms of the numerical underpinning of
372 methods applied. Indeed, many such studies attempt to derive provenance estimates
373 only qualitatively (e.g. Fitzsimmons et al, 2009), or, in the few recent cases where
374 robust unmixing models have been applied, relatively simple approaches to
375 incorporating uncertainty into models have been taken (e.g. Liu et al., 2016).

376 The successful application of a Bayesian model within an MCMC framework to
377 aeolian sands of a small erg in this study demonstrates the potential of this approach
378 more widely. It is particularly likely to complement detrital zircon U-Pb dating
379 provenance studies, and may prove especially useful in settings where there are
380 insufficient zircon grains to enable the application of this method (e.g. Jia et al.,
381 2015, Nie and Peng, 2014, Ren et al., 2014, Thorpe et al., 1992). Further application
382 of the methods demonstrated here is required to test the ability of such methods
383 globally, but these results suggest a promising future, and a new direction for aeolian
384 provenance studies. For instance, it would be useful to explore the power of these
385 methods in larger-scale settings, such as the continental dunefields of southern
386 Africa and Australia, where provenance studies have been used to explore the

relationship between tectonic setting and sedimentation (e.g. Garzanti et al., 2014) and explore the long-term evolution of landscapes (e.g. Pell et al., 1997; 2000). This will also elucidate the importance of diversity of local geology and weathering regimes in producing sufficiently distinctive fingerprints.

Accurate propagation of the uncertainties associated with the component contributions is also a valuable aspect of the methodology employed here and allows more realistic interpretation of the data. For instance, the most abundant two source components of samples E ($Qt_2 = 62.3\%$; $Qc = 23\%$) and D ($Egm = 56.3\%$; $Qc = 28.7\%$) might suggest similar proportions of the major components at these dunes; roughly 60:25. However, consideration of the lower confidence of the fingerprint of sample D (Figure 6) reveals that whilst the composition of this sample is much more open to interpretation, sample E is quite clearly dominated by the alluvial fan-derived sands (Qt_2).

400

401 4.3 Implications for aeolian sediment transport pathways

Overall, the surrounding Quaternary fans and terraces contribute most (~45%) to the composition of Ashkzar erg; yet this is a disproportionately low value, given that they represent 78% of the surrounding area. Conversely, the size of the overall contributions from the Quaternary clay flats (~26%) and Eocene marls (~28%) to the samples studied reveals the importance of these landscape units as sediment sources, given that these units occupy only 13% and 7% of the surrounding area, respectively. The importance of the marls as a source sediment, which outcrop only to the north of the Ashkzar dunefield, suggests that net wind regime alone cannot be considered as indicative of the net sediment transport in the region (Figure 1), as

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

411 westerly winds are equally strong here yet import much less sediment. Both the wind
412 regime and potential sediment sources must be considered when evaluating net
413 aeolian sediment flux.

414 There is much spatial variation in the composition of the dune sands of Ashkzar erg
415 (Figure 6). Even before the geochemical composition of these sands is considered,
416 such variability is evident from the differing grain size profiles in the eight samples
417 investigated here (Table 1). Sample G, from the far south of the dunefield, contains
418 ~20% coarse sands (defined here as $> 600 \mu\text{m}$), an unusually high figure for an
419 aeolian dune, although this sample is taken close to the border with the mapped
420 region of slipfaceless dome dunes, which tend to accumulate from coarser sands
421 (Lancaster, 1995). That said, sample H, from within the dome dunes, is not unusually
422 coarse.

423 Broadly, and considering the uncertainties presented by the methodology proposed
424 herein, two groups can be discerned within the samples geochemically analysed for
425 provenance (Figure 6). Samples taken from along the south and west of the
426 dunefield (B, E, F and G) are dominated to varying degrees by sediment from the
427 surrounding Quaternary fans and terraces (i.e. source unit Qt2), whereas most
428 samples to the north and east (A, D and H) show much greater contributions from
429 the Eocene marls (Egm) and clay flats (Qc). However, the division is not clear-cut;
430 sample C, in the northeast of the dunefield, has a dominant component from the Qt2
431 unit (with the second component only very slightly overlapped at 2σ confidence
432 levels). It is, perhaps, unsurprising that the more northerly samples tend to show an
433 increased input from the Eocene marls (Egm), as these units have been shown to
434 contribute disproportionately to the sediment flux in the area, and also outcrop
435 exclusively on the northern side of the valley (Figure 1).

1
2
3 436 The provenance of the sands is even less readily correlated with dune morphology,
4
5 437 with the Qt2-dominated sands occurring within three defined dune morphological
6
7 438 zones (barchans, barchanoid ridges and asymmetrical barchans). Samples A and B,
8
9 439 the closest pair of samples studied (~2.2 km apart), yield very different provenance
10
11 440 fingerprints, despite both lying within the region of Ashkzar erg dominated by
12
13 441 barchanoid ridges. The overall morphology of the dunes (Figure 3) supports spatially
14
15 442 and /or temporally variable sediment availability, with the transformation of barchans
16
17 443 to barchanoid ridges essentially being sediment-supply controlled, and asymmetric
18
19 444 barchan/linear forms believed to be the result of asymmetries of sediment supply, or
20
21 445 changes to the wind regime (Bagnold, 1941; Lancaster 1995).
22
23
24
25

26 446 The heterogeneity in the sediment provenance evident here suggests that either a)
27
28 447 some kind of fractionation of the aeolian sediment flux is occurring, with different
29
30 448 sources depositing sediment at different locations or b) different sediment transport
31
32 449 pathways have been active intermittently and asynchronously during the formation of
33
34 450 the dunefield. The similar physical composition (i.e. grain size) of the sources, and
35
36 451 the lack of evidence of systematic variation across the dunefield, would tend to
37
38 452 support the latter suggestion. Different sediment pathways might result from different
39
40 453 sources become more or less active over time, or might result from changing wind
41
42 454 regime over long (i.e. late Quaternary) timescales. The heterogeneity evident also
43
44 455 suggests that during dune accumulation periods, large-scale mixing of aeolian sands
45
46 456 from different sources (which might be expected given transport distances of 10-50
47
48 457 km) is not occurring. In the absence of any chronological control for these dunes,
49
50 458 such hypotheses cannot be conclusively tested currently, but establishing the
51
52 459 relative roles of spatial and temporal variability in dune accumulation would be a
53
54 460 worthwhile exercise.
55
56
57
58
59
60

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

461

462

463
464
465
466
467
468
469
470
471
472
473
474
475
476
477
478

479

480
481
482
483
484
485

5. Conclusion

In dryland environments, understanding the main sources for aeolian sediments is an essential step in developing management strategies to reduce aeolian sediment loadings and wind erosion. Establishing aeolian sediment pathways, however, is not usually straightforward and is complicated when multiple potential source areas might contribute to a region of net sand accumulation. The method proposed here, based on methodologies applied to fluvial sediments, uses a suite of geochemical data to identify the most apposite characteristics (the ‘fingerprint’) for discerning superficially similar sources of aeolian sediment. Whereas these methods have become widely adopted in fluvial geomorphology and catchment science over the past two decades, they remain almost unused in aeolian science. Here, it has been successfully demonstrated on fine sand in a small dunefield in central Iran, but it might be applied equally to dust (i.e. silt) flux, although longer transport distances are liable to prove more difficult to fingerprint unless relatively discrete and distinct sources can be identified. The use of MCMC methods to provide confidence estimates in the mixing model output enables more rigorous interpretation of the relative importance of different sediment sources.

This method revealed within Ashkzar erg unexpected spatial heterogeneity of dune composition (and thus provenance), which has a complex relationship with the position within the dunefield, the dune type and other physical characteristics. The Eocene marls in the surrounding area have been shown to contribute disproportionately to the sediments of the dunes. In terms of management of sand and dust hazard at this location, both the original source areas and those parts of the

dunefield enriched in the Egm component might be viewed as priority targets for landscape stabilization efforts, due to their apparent propensity for aeolian mobilization.

More widely, the methods proposed here for aeolian provenance unmixing method can be applied to any mixed-source aeolian sediment to elucidate differing susceptibilities to aeolian deflation, and reveal transport pathways at timescales longer than those possible by either field study or remote sensing. Disciplines which might benefit from the adoption of such methods include not just aeolian geomorphology, but also dryland land management, soil science, engineering geology and potentially palaeoenvironmental and palaeoclimatological studies. A combination of the methods presented herein with geochronological studies may enable calculation of flux rates to provide quantification of long-term sediment fluxes, even when, as is very often the case with aeolian sediments, transport pathways are complex and multi-phase.

References

- Amiraslani, F. and Dragovich, D. (2011). Combating desertification in Iran over the last 50 years: An overview of changing approaches. *Journal of Environmental Management* 92 (1), 1-13. doi: 10.1016/j.jenvman.2010.08.012.
- Bagnold, R.A. 1941. The physics of blown sand. Methuen, London.
- Box, G.E.P & Cox, D.R. (1964). An analysis of transformations. *Journal of the Royal Statistical Society, Series B*. 26 (2): 211–252.

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

510 Brewer, M. J., Filipe, J. A. N., Elston, D. A., Dawson, L. A., Mayes, R. W., Soulsby,
511 Ch., & Dunn, S. M. (2005). A Hierarchical Model for Compositional Data
512 Analysis. *Journal of Agricultural, Biological, and Environmental Statistics*, 10(1),
513 19–34. doi:10.1198/108571105X28200

514 Chen, F., Fang, N., & Shi, Z. (2016). Using biomarkers as fi ngerprint properties to
515 identify sediment sources in a small catchment. *Science of the Total*
516 *Environment*, 557-558, 123–133. doi:10.1016/j.scitotenv.2016.03.028

517 Collins, A. L., & Walling, D. E. (2007). Sources of fine sediment recovered from the
518 channel bed of lowland groundwater-fed catchments in the UK. *Geomorphology*,
519 88, 120–138. doi:10.1016/j.geomorph.2006.10.018

520 Collins, A. L., Zhang, Y., McChesney, D., Walling, D. E., Haley, S. M., & Smith, P.
521 (2012). Sediment source tracing in a lowland agricultural catchment in southern
522 England using a modified procedure combining statistical analysis and
523 numerical modelling. *Science of the Total Environment*, 414, 301–317.
524 doi:10.1016/j.scitotenv.2011.10.062

525 Collins, A. L., Zhang, Y. S., Duethmann, D., Walling, D. E., & Black, K. S. (2013).
526 Using a novel tracing-tracking framework to source fine-grained sediment loss to
527 watercourses at sub-catchment scale. *Hydrological Processes*, 27(6), 959–974.
528 doi:10.1002/hyp.9652

529 Collins, A. L., Zhang, Y., Walling, D. E., Grenfell, S. E., & Smith, P. (2010). Tracing
530 sediment loss from eroding farm tracks using a geochemical fingerprinting
531 procedure combining local and genetic algorithm optimisation. *Science of the*
532 *Total Environment*, 408(22), 5461–5471. doi:10.1016/j.scitotenv.2010.07.066

- 533 Collins, A. L., Zhang, Y., Walling, D. E., Grenfell, S. E., Smith, P., Grischeff, J., ...
534 Brogden, D. (2012). Quantifying fine-grained sediment sources in the River Axe
535 catchment, southwest England: Application of a Monte Carlo numerical
536 modelling framework incorporating local and genetic algorithm optimisation.
537 *Hydrological Processes*, 26(13), 1962–1983. doi:10.1002/hyp.8283
- 538 Collins, A.L., Pulley, S., Foster, I.D.L., Gellis A., Porto, P., Horowitz, A.J. (2016).
539 Sediment source fingerprinting as an aid to catchment management: A review of
540 the current state of knowledge and a methodological decision-tree for end-
541 users. *Journal of Environmental Management*, 194, 86–108.
542 doi.org/10.1016/j.jenvman.2016.09.075
- 543 Cooper RJ and Krueger T (2017, in press). An extended Bayesian sediment
544 fingerprinting mixing model for the full Bayes treatment of geochemical
545 uncertainties. *Hydrological Processes*. DOI: 10.1002/hyp.11154.
- 546 Cooper, R. J., Krueger, T., Hiscock, K. M., & Rawlins, B. G. (2014). Sensitivity of
547 fluvial sediment source apportionment to mixing model assumptions: A
548 Bayesian model comparison. *Water Resources Research*, 9031–9047.
549 doi:10.1002/2014WR016194.
- 550 Cooper, R. J., Krueger, T., Hiscock, K. M., & Rawlins, B. G. (2015). High-temporal
551 resolution fluvial sediment source fingerprinting with uncertainty: A Bayesian
552 approach. *Earth Surface Processes and Landforms*, 40(1), 78–92.
553 doi:10.1002/esp.3621
- 554 Daga, R., Ribeiro Guevara, S., Sánchez, M. L., & Arribére, M. (2008). Source
555 identification of volcanic ashes by geochemical analysis of well preserved

- lacustrine tephra in Nahuel Huapi National Park. *Applied Radiation and Isotopes*, 66(10), 1325–1336. doi:10.1016/j.apradiso.2008.03.009
- Defant, M.J. and Drummond, M.S. 1990. Derivation of some modern arc magmas by melting of young subducted lithosphere. *Nature* 367, 662–665
- Delmonte, B., Baroni, C., Andersson, P.S., Schoberg, H., Hansson, M., Aciego, S., Petit, J.-R., Albani, S., Mazzola, C., Maggi, V., Frezzotti, M., 2010. Aeolian dust in the Talos Dome ice core (East Antarctica, Pacific/Ross Sea sector): Victoria Land versus remote sources over the last two climate cycles. *Journal of Quaternary Science* 25, 1327–1337.
- Delmonte, B., Basile-Doelsch, I., Petit, J.R., Maggi, V., Revel-Rolland, M., Michard, A., Jagoutz, E., Grousset, F., 2004. Comparing the Epica and Vostok dust records during the last 220,000 years: stratigraphical correlation and provenance in glacial periods. *Earth-Science Reviews* 66, 63–87.
- Dou, Y., Yang, S., Liu, Z., Clift, P. D., Shi, X., Yu, H., & Berne, S. (2010). Provenance discrimination of siliciclastic sediments in the middle Okinawa Trough since 30ka: Constraints from rare earth element compositions. *Marine Geology*, 275(1–4), 212–220. doi:10.1016/j.margeo.2010.06.002
- Douglas, G., Caitcheon, G., & Palmer, M. (2009). Sediment source identification and residence times in the Maroochy River estuary, southeast Queensland, Australia. *Environmental Geology*, 57(3), 629–639. doi:10.1007/s00254-008-1336-7
- Egozcue, J. J., Pawlowsky-Glahn, V., Mateu-Figueras, G., & Barceló-Vidal, C. (2003). Isometric Logratio Transformations for Compositional Data Analysis.

- 1
2
3 579 *Mathematical Geology*, 35(3), 279–300. doi:10.1023/A:1023818214614
4
5
6 580 Fox, J. F., & Papanicolaou, A. N. (2008). An un-mixing model to study watershed
7
8 581 erosion processes. *Advances in Water Resources*, 31, 96–108.
9
10 582 doi:10.1016/j.advwatres.2007.06.008
11
12
13 583 Garzanti, E., Vermeesch, P., Ando, S., Vezzoli, G., Valagussa, M., Allen, K., Kadi,
14
15 584 K.A., Al-Juboury, A.I.A., 2013. Provenance and recycling of Arabian desert
16
17 585 sand. *Earth-Science Reviews*. 120, 1-19.
18
19
20
21 586 Garzanti, E., Vermeesch, P., Padoan, M., Resentini, A., Vezzoli, G., Ando, S., 2014.
22
23 587 Provenance of Passive-Margin Sand (Southern Africa). *Journal of Geology*,
24
25 588 122(1), 17-42. doi:10.1086/674803
26
27
28 589 Gellis, A. C., & Noe, G. B. (2013). Sediment source analysis in the Linganore Creek
29
30 590 watershed, Maryland, USA, using the sediment fingerprinting approach: 2008 to
31
32 591 2010. *Journal of Soils and Sediments*, 13(10), 1735–1753. doi:10.1007/s11368-
33
34 592 013-0771-6
35
36
37
38 593 Haddadchi, A, Ryder, D.S., Evrard, O., Olley, J. (2013). Sediment fingerprinting in
39
40 594 fluvial systems: review of tracers, sediment sources and mixing models. *Int. J.*
41
42 595 *Sediment Res.*, 28, 560–578
43
44
45 596 Huntsman-Mapila, P., Kampunzu, A.B., Vink, B., Ringrose, S., 2005. Cryptic
46
47 597 indicators of provenance from the geochemistry of the Okavango Delta
48
49 598 sediments, Botswana. *Sedimentary Geology* 174, 123-148.
50
51
52
53 599 Honda, M., Yabuki, S., & Shimizu, H. S. H. I. (2004). Geochemical and isotopic
54
55 600 studies of aeolian sediments in China. *Sedimentology*, 211–230.
56
57 601 doi:10.1046/j.1365-3091.2003.00618.x
58
59
60

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

602 Jia, Y., Fu, B., Jolivet, M. and Zheng, S. Cenozoic tectono-geomorphological growth
603 of the SW Chinese Tian Shan: insight from AFT and detrital zircon U-Pb data.
604 *Journal of Asian Earth Sciences*, 2015, 111, 395-413.

605 Koiter, A. J., Owens, P. N., Petticrew, E. L., & Lobb, D. A. (2013). The behavioural
606 characteristics of sediment properties and their implications for sediment
607 fingerprinting as an approach for identifying sediment sources in river basins.
608 *Earth-Science Reviews*, 125, 24–42. doi:10.1016/j.earscirev.2013.05.009

609 Lancaster, N. 1995. Geomorphology of Desert Dunes. Routledge, London.

610 Lin, J., Huang, Y., Wang, M. kuang, Jiang, F., Zhang, X., & Ge, H. (2015). Assessing
611 the sources of sediment transported in gully systems using a fingerprinting
612 approach: An example from South-east China. *Catena*, 129, 9–17.
613 doi:10.1016/j.catena.2015.02.012

614 Liu, B.L., Niu, Q.H., Qu, J.J., Zu, R.P. (2016). Quantifying the provenance of aeolian
615 sediments using multiple composite fingerprints. *Aeolian Research*, 22, 117-
616 122. doi:10.1016/j.aeolia.2016.08.002

617 Lunn, D.J., Thomas, A., Best, N., and Spiegelhalter, D. (2000) WinBUGS -- a
618 Bayesian modelling framework: concepts, structure, and extensibility. *Statistics
619 and Computing*, 10:325--337.

620 Martínez-Carreras, N., Udelhoven, T., Krein, A., Gallart, F., Iffly, J. F., Ziebel, J. and
621 Walling, D. E. (2010). The use of sediment colour measured by diffuse
622 reflectance spectrometry to determine sediment sources: Application to the
623 Attert River catchment. *Journal of Hydrology*, 382(1-4), 49–63.
624 doi:10.1016/j.jhydrol.2009.12.017

- 625 Massoudieh, A., Gellis, A., Banks, W. S., & Wieczorek, M. E. (2013). Suspended
626 sediment source apportionment in Chesapeake Bay watershed using Bayesian
627 chemical mass balance receptor modeling. *Hydrological Processes*, 27(24),
628 3363–3374. doi:10.1002/hyp.9429
- 629 Motha, J. A., Wallbrink, P. J., Hairsine, P. B., & Grayson, R. B. (2003). Determining
630 the sources of suspended sediment in a forested catchment in southeastern
631 Australia. *Water Resources Research*, 39(3), 1056. doi:10.1029/2001wr000794
- 632 Muhs, D. R., Lancaster, N. and Skipp, G.L. (2017). A complex origin for the Kelso
633 Dunes, Mojave National Preserve, California, USA: A case study using a simple
634 geochemical method with global applications. *Geomorphology*, 276, 222-243.
635 doi: 10.1016/j.geomorph.2016.10.002
- 636 Mukundan, R., Walling, D. E., Gellis, A. C., Slattery, M. C., & Radcliffe, D. E. (2012).
637 Sediment Source Fingerprinting: Transforming From a Research Tool to a
638 Management Tool. *Journal of the American Water Resources Association*,
639 48(6), 1241–1257. doi:10.1111/j.1752-1688.2012.00685.x
- 640 Naddafi, K., Nabizadeh, R., Soltanianzadeh, Z., Ehrampoosh, M.H., 2006.
641 Evaluation of dustfall in the air of Yazd. *Journal of Environmental Health*
642 *Science and Engineering*, 3, 161-168.
- 643 Nanson, G.C., Chen, X.Y., Price, D.M. (1995). Aeolian and fluvial evidence of
644 changing climate and wind patterns during the past 100 Ka in the Western
645 Simpson Desert, Australia. *Palaeogeography Palaeoclimatology Palaeoecology*
646 113, 87-102.
- 647 Nie, J. and Peng, W. (2014). Automated SEM–EDS heavy mineral analysis reveals

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

648 no provenance shift between glacial loess and interglacial paleosol on the
649 Chinese Loess Plateau. *Aeolian Research* 13, 71-75.

650 Nosrati, K., Govers, G., Semmens, B. X., & Ward, E. J. (2014). A mixing model to
651 incorporate uncertainty in sediment fingerprinting. *Geoderma* 217-218, 173–180.
652 doi:10.1016/j.geoderma.2013.12.002

653 Owens, P.N., Blake, W.H., Gaspar, L., Gateuille, D., Koiter, A.J., Lobb, D.A.,
654 Petticrew, E.L., Reiffarth, D.G., Smith, H.G. & Woodward, J.C. (2016).
655 Fingerprinting and tracing the sources of soils and sediments: Earth and ocean
656 science, geoarchaeological, forensic, and human health applications. *Earth-*
657 *Science Reviews*, 162, 1-23. doi: 10.1016/j.earscirev.2016.08.012

658 Palmer, M. J., & Douglas, G. B. (2008). A Bayesian statistical model for end member
659 analysis of sediment geochemistry, incorporating spatial dependences. *Journal*
660 *of the Royal Statistical Society. Series C: Applied Statistics* 57(3), 313–327.
661 doi:10.1111/j.1467-9876.2007.00615.x

662 Parnell, A. C., Phillips, D. L., Bearhop, S., Semmens, B. X., Ward, E. J., Moore, J.
663 W., Inger, R. (2013). Bayesian stable isotope mixing models. *Environmetrics*
664 24(6), 387–399. doi:10.1002/env.2221

665 Pell, S.D., Williams, I.S., Chivas, A.R., 1997. The use of protolith zircon-age
666 fingerprints in determining the protosource areas for some Australian dune
667 sands. *Sedimentary Geology* 109, 233-260.

668 Pell, S.D., Chivas, A.R., Williams, I.S. 2000. The Simpson, Strzelecki and Tirari
669 Deserts: development and sand provenance. *Sedimentary Geology* 130 (1-2),
670 107–130. doi: 10.1016/S0037-0738(99)00108-6

- 671 Pethierick, L., McGowan, H., Moss, P., 2008. Climate variability during the Last
672 Glacial Maximum in eastern Australia: evidence of two stadials? *Journal of*
673 *Quaternary Science* 23, 787-802.
- 674 Pittam, N. J., Foster, I. D. L., & Mighall, T. M. (2009). An integrated lake-catchment
675 approach for determining sediment source changes at Aqualate Mere, Central
676 England. *Journal of Paleolimnology*, 42(2), 215–232. doi:10.1007/s10933-008-
677 9272-9
- 678 Rao, W., Tan, H., Jiang, S., & Chen, J. (2011). Trace element and REE
679 geochemistry of fine- and coarse-grained sands in the Ordos deserts and links
680 with sediments in surrounding areas. *Chemie Der Erde - Geochemistry*, 71(2),
681 155–170. doi:10.1016/j.chemer.2011.02.003
- 682 Ren, R., Han, B-F., Xu, Z. and Li, Q. 2014. When did the subduction first initiate in
683 the southern Paleo-Asian Ocean: New constraints from a Cambrian intra-
684 oceanic arc system in West Junggar, NW China. *Earth and Planetary Science*
685 *Letters* 388, 222–236
- 686 Semmens, B. X., Moore, J. W., & Ward, E. J. (2009). Improving Bayesian isotope
687 mixing models: A response to Jackson et al. (2009). *Ecology Letters*, 12(3), 10–
688 12. doi:10.1111/j.1461-0248.2009.01283.x
- 689 Sherriff, S. C., Franks, S. W., Rowan, J. S., Fenton, O., & Ó'hUallacháin, D. (2015).
690 Uncertainty-based assessment of tracer selection, tracer non-conservativeness
691 and multiple solutions in sediment fingerprinting using synthetic and field data.
692 *Journal of Soils and Sediments*, 15(10), 2101–2116. doi:10.1007/s11368-015-
693 1123-5

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

694 Smith, H. G., & Blake, W. H. (2014). Sediment fingerprinting in agricultural
695 catchments: A critical re-examination of source discrimination and data
696 corrections. *Geomorphology*, 204, 177–191.
697 doi:10.1016/j.geomorph.2013.08.003

698 Stewart, H. A., Massoudieh, A., & Gellis, A. (2015). Sediment source apportionment
699 in Laurel Hill Creek, PA, using Bayesian chemical mass balance and isotope
700 fingerprinting. *Hydrological Processes*, 29(11), 2545–2560.
701 doi:10.1002/hyp.10364

702 Stone, M., Collins, A. L., Silins, U., Emelko, M. B., & Zhang, Y. S. (2014). The use of
703 composite fingerprints to quantify sediment sources in a wildfire impacted
704 landscape, Alberta, Canada. *Science of the Total Environment*, 473-474, 642–
705 650. doi:10.1016/j.scitotenv.2013.12.052

706 Thorpe, R.I., Hickman, A.H., Davis, D.W., Mortensen, J.K. and Trendall, A.F., 1992.
707 U/Pb zircon geochronology of Archaean felsic units in the Marble Bar region,
708 Pilbara Craton, Western Australia. *Precambrian Research*, 56, 169-189.

709 Vale, S. S., Fuller, I. C., Procter, J. N., Basher, L. R., & Smith, I. E. (2016).
710 Characterization and quantification of suspended sediment sources to the
711 Manawatu River, New Zealand. *Science of The Total Environment*, 543, 171–
712 186. doi:10.1016/j.scitotenv.2015.11.003

713 Voli, M. T., Wegmann, K. W., Bohnenstiehl, D. R., Leithold, E., Osburn, C. L., &
714 Polyakov, V. 2013. Fingerprinting the sources of suspended sediment delivery
715 to a large municipal drinking water reservoir: Falls Lake, Neuse River, North
716 Carolina, USA. *Journal of Soils and Sediments*, 13(10), 1692–1707.

- doi:10.1007/s11368-013-0758-3
- Walling, D.E., Collins, A.L., & Stroud, R.W. 2008. Tracing suspended sediment and particulate phosphorus sources in catchments. *Journal of Hydrology*, 350(3-4), 274–289. doi:10.1016/j.jhydrol.2007.10.047
- Walling, D.E. 2013. The evolution of sediment source fingerprinting investigations in fluvial systems. *Journal of Soils and Sediments*, 13(10), 1658-1675. doi: 10.1007/s11368-013-0767-2
- Wasklewicz, T.A., Meek, N., 1995. Provenance of aeolian sediment: The upper Coachella Valley, California. *Physical Geography* 16, 539-556.
- Wilkinson, S.N., Hancock, G.J., Bartley, R., Hawdon, A.A., & Keen, R.J. (2013). Using sediment tracing to assess processes and spatial patterns of erosion in grazed rangelands, Burdekin River basin, Australia. *Agriculture, Ecosystems and Environment*, 180, 90–102. doi:10.1016/j.agee.2012.02.002
- Wilson, C.G., Papanicolaou, A.N.T., & Denn, K.D. (2012). Partitioning fine sediment loads in a headwater system with intensive agriculture. *Journal of Soils and Sediments*, 12(6), 966–981. doi:10.1007/s11368-012-0504-2
- Winspear, N.R., Pye, K., 1996. Textural, geochemical and mineralogical evidence for the sources of aeolian sand in central and southwestern Nebraska, USA. *Sedimentary Geology* 101, 85-98.
- Yang, X., Liu, Y., Li, C., Song, Y., Zhu, H., Jin, X., 2007. Rare earth elements of aeolian deposits in Northern China and their implications for determining the provenance of dust storms in Beijing. *Geomorphology* 87, 365-377.

1
2
3 739
4
5
6 740
7
8
9 741
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

For Peer Review

742 Tables

743 Table 1

		Grain size (μm)					
		<62.5	62.5 - 150	150 - 300	300 - 600	600 - 1180	1180 - 1700
Mean Eocene marl (Egm) source (% $\pm 1\sigma$)		3.0 \pm 1.4	40.2 \pm 10.6	20.0 \pm 6	15.1 \pm 4.2	15.6 \pm 6.7	6.1 \pm 5
Mean Quaternary clay flat (Qc) source (% $\pm 1\sigma$)		3.1 \pm 1.3	29.4 \pm 9.2	21.8 \pm 5.6	19.0 \pm 3.9	19.7 \pm 8.1	6.9 \pm 4
Mean Quaternary terrace/fan (Qt2) source (% $\pm 1\sigma$)		2.2 \pm 2.3	34.7 \pm 7.3	20.4 \pm 5.6	17.1 \pm 3.9	19.3 \pm 5.3	6.4 \pm 2
Mean Ashkzar erg dune sands (% \pm 1 σ)		1.1 \pm 0.9	47.8 \pm 9.3	29.7 \pm 8.4	17.4 \pm 13.9	3.4 \pm 5.4	0.6 \pm 1.3
Ashkzar erg dune samples (%)	A	0.3	57.6	38.2	3.4	0.5	0.0
	B	0.5	41.0	18.1	39.0	1.4	0.0
	C	2.1	43.9	16.7	35.9	1.4	0.0
	D	0.5	42.5	34.0	22.0	1.0	0.0
	E	2.7	51.8	31.2	7.9	5.0	1.4
	F	1.0	64.5	32.0	2.5	0.0	0.0
	G	1.0	36.8	28.0	14.4	16.2	3.6
	H	0.6	44.6	39.3	13.7	1.8	0.0

744

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

745 Table 2

Fingerprint property	Chi square	p value	Fingerprint property	Chi square	p-value
La	6.669	0.036 [*]	Y	4.151	0.125
Ce	7.476	0.024 ^{**}	Zr	3.744	0.154
Pr	9.552	0.008 ^{**}	Nb	0.582	0.748
Nd	0.415	0.813	Hf	16.1	<0.001 ^{***}
Sm	0.081	0.96	Ta	4.922	0.085 ^{**}
Eu	10.23	0.006 ^{**}	Th	10.28	0.006 ^{**}
Gd	0.434	0.805	U	4.8	0.091
Tb	0.017	0.992	As	5.166	0.076
Dy	1.359	0.507	Bi	0.725	0.695
Ho	8.067	0.018 [*]	Cd	0.111	0.946
Er	9.257	0.01 [*]	Ga	13.4	<0.001 ^{***}
Tm	8.373	0.015 [*]	Ge	0.59	0.745
Yb	1.026	0.599	In	1.215	0.545
Lu	0.104	0.949	Li	7.213	0.027 [*]
ΣREE	7.086	0.029 [*]	Mo	0.227	0.893
Eu/Eu [*]	10.23	0.006 ^{**}	P	13.05	<0.001 ^{***}
(Nd/Yb)	0.785	0.675	S	16.36	<0.001 ^{***}
(Gd/Yb) _n	0.729	0.695	Sb	1.409	0.494
(La/Yb) _n	13.89	0.001 ^{**}	Se	0.858	0.651
(La/Sm) _n	4.725	0.094 [*]	Sn	6.466	0.039 [*]
(La/Lu) _n	12.78	0.002 ^{**}	Te	4.685	0.096
δCe	5.435	0.041 [*]	Ti	6.56	0.038 [*]
Rb	15.18	<0.001 ^{***}	Tl	6.614	0.037 [*]
Sr	16.44	<0.001 ^{***}	W	0.086	0.958
Ba	6.379	0.041 [*]	Mn	1.673	0.433
V	3.083	0.214	Si	0.342	0.843
Cr	5.359	0.069	¹⁴³ Nd	1.042	0.534
Co	1.271	0.53	¹⁴⁴ Nd	2.031	0.362
Ni	3.998	0.135	⁸⁶ Sr	5.754	0.056
Cu	3.18	0.204	⁸⁷ Sr	7.124	0.028 [*]
Zn	0.236	0.889			

746

747 Table 3

Sediment	Tracer	Optimum composite fingerprints					
		Rb	Sr	⁸⁷ Sr	(La/Yb) _n	Ga	δCe
Sand dune	Mean	7.8	144	85	7.1	1.2	0.69
	SD	0.74	18.6	28	0.54	0.11	0.020
Source	Tracer	Rb	Sr	⁸⁷ Sr	(La/Yb) _n	Ga	δCe
Egm	Mean	8.9	293	82	6.9	1.3	0.70
	SD	2.3	192	31	0.58	0.29	0.085
Qc	Mean	10	139	91	7.4	1.1	0.69
	SD	2.6	36.5	31	0.54	0.27	0.071
Qt2	Mean	7.1	163	98	7.7	0.99	0.74
	SD	0.82	135	44	0.89	0.26	0.27

748

749 Table 4

	Source	Tracer					
		Rb	Sr	⁸⁷ Sr	(La/Yb) _n	Ga	δCe
Raw data	Egm	0.944	0.362	0.930	0.996	0.980	0.810
	Qc	0.347	0.734	0.486	0.794	0.900	0.167
	Qt2	0.983	0.019	0.182	0.557	0.710	0.013
	Source	Tracer					
		Rb	Sr	⁸⁷ Sr	(La/Yb) _n	Ga	δCe
Box-Cox transformed data	Egm	0.944	0.724	0.930	0.996	0.980	0.978
	Qc	0.347	0.931	0.486	0.794	0.900	0.542
	Qt2	0.983	0.085	0.182	0.557	0.710	0.061

750

751 Table 5

752

Sediment samples	Source	Mean (%)	SD (%)	MC error	Median	Percentile (2.5)	Percentile (97.5)
A	Egm	44.1	11.5	0.003	44.5	21	64.7
	Qc	39.8	11.2	0.003	37.9	22.7	65.8
	Qt2	16.2	8.3	0.001	15.2	3.6	34
B	Egm	32.6	7.3	0.002	33	18.2	46.2
	Qc	5.1	3.8	0.001	4.1	0.6	15
	Qt2	62.2	6	0.001	62.1	50.3	73.7
C	Egm	20.8	6.3	0.002	21	8.1	33
	Qc	27.5	6.5	0.002	26.6	17.1	43
	Qt2	51.6	5.2	0.000	51.1	42	62.5
D	Egm	56.3	13	0.004	56.9	29.9	79.4
	Qc	28.7	10.5	0.003	27	12.2	53.4
	Qt2	14.9	9.4	0.001	14.4	0.4	34.2
E	Egm	14.3	5	0.001	14.3	4.6	24.6

F	Qc	23	5.3	0.001	22.3	14.2	35.1
	Qt2	62.6	4.8	0.000	62.7	52	71.9
	Egm	17.6	6.3	0.001	17.7	4.8	29.9
G	Qc	32.4	7.4	0.002	31.5	20.4	49.6
	Qt2	49.9	5.5	0.001	49.4	39.7	61.5
	Egm	3.4	3.2	0.000	2.44	0.000	12.2
H	Qc	3.9	3.9	0.000	2.6	0.000	14.7
	Qt2	92.7	6	0.001	94	78.2	99.7
	Egm	37.3	11.2	0.003	37.8	14.5	57.9
	Qc	49.7	12.6	0.004	47.7	30	79
	Qt2	13	8.6	0.001	12	0.4	31.5

753

754

For Peer Review

Figure captions

756

Figure 1: Location and geological map of the Yazd-Ardekan Plain and sampling sites. Dominant and minor wind directions shown in Part C. Number of sampling points = 57. Qt2, Qc, Egm (potential sediment sources) and Qsd (sediment) represent young alluvial fans and terraces, clay flats, gypsiferous marl and sand dunes, respectively.

762

Figure 2: Source and sediment sink regions within the study area. Source regions include: a) clay flats (Qc); b) gypsiferous marl (Egm); and c) young alluvial fans and terraces (Qt2). Sediment sinks include: d) sand dunes (Qsd).

766

Figure 3. Morphological mapping of dune types within Ashkzar erg reveals the dominance of barchans and barchanoid ridges. There are less distinct zones within the dunefield in the north, where the interdunes are sandy and the transverse forms much less distinct, and in the far southeast, where patchy slipface-less dunes dominate. Base imagery is courtesy of Google Earth™, and letters refer to the eight samples analysed for physical and geochemical characteristics within the dunefield.

773

Figure 4: A directed acyclic graph of the Bayesian mixing model employed in this study.

776

Figure 5: Two-dimensional scatter plot of the first and second discriminant functions from stepwise DFA for the source groups Egm (Eocene gypsiferous marls), Qc (Quaternary clay flats) and Qt2 (Quaternary alluvial fans and terraces).

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

Figure 6: Source contributions for each aeolian sediment samples by Bayesian mixing model with 95% credible limits. Base imagery is courtesy of Google Earth™.

Table captions

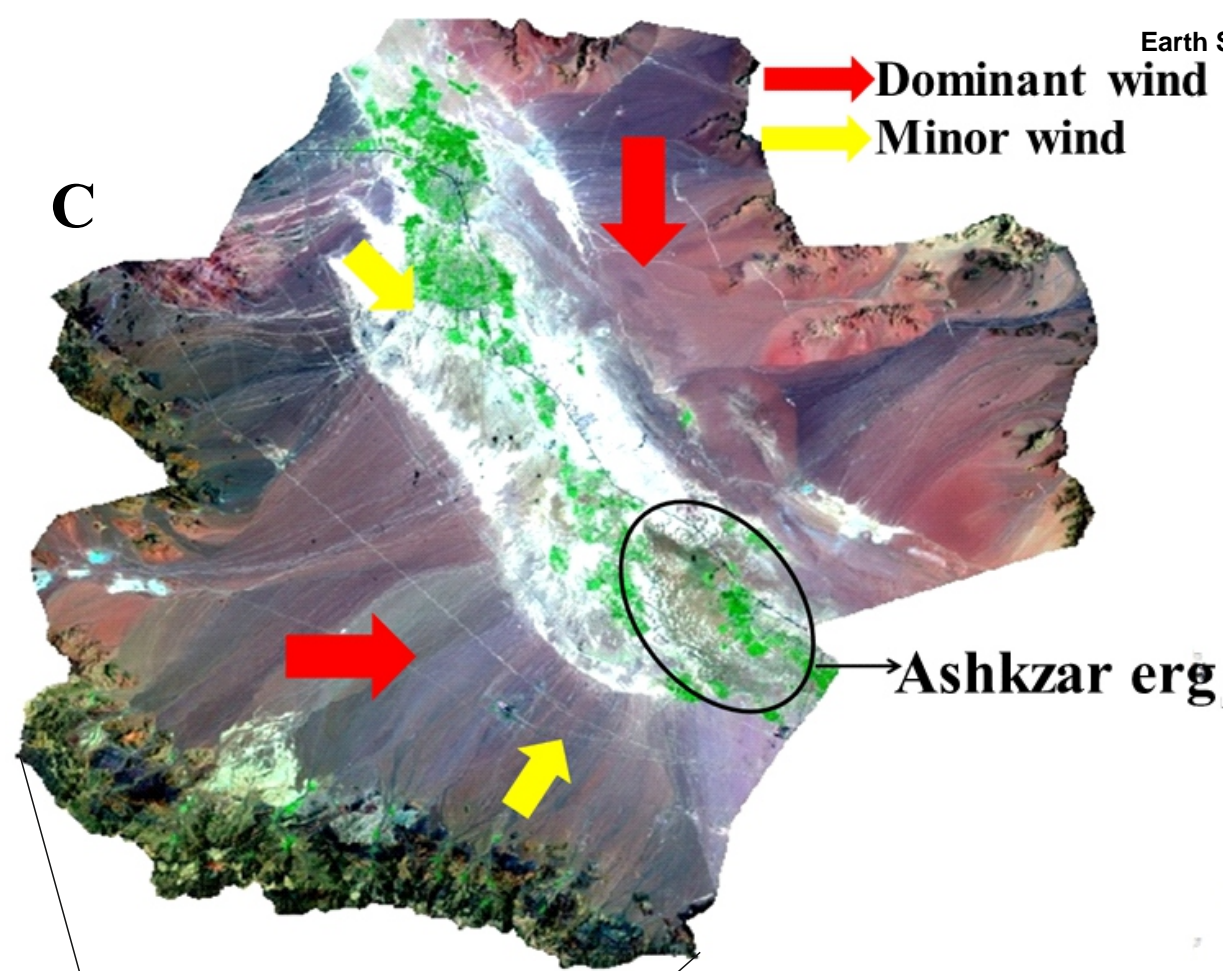
Table 1: Grain size data for the source areas, the dunefield, and individual samples from within the dunefield.

Table 2: Kruskal-Wallis H test results for selecting fingerprint properties for distinguishing individual source types. Confidence is highlighted at >95% with a single asterisk, >99% with a double asterisk, and >99.9% with a triple asterisk.

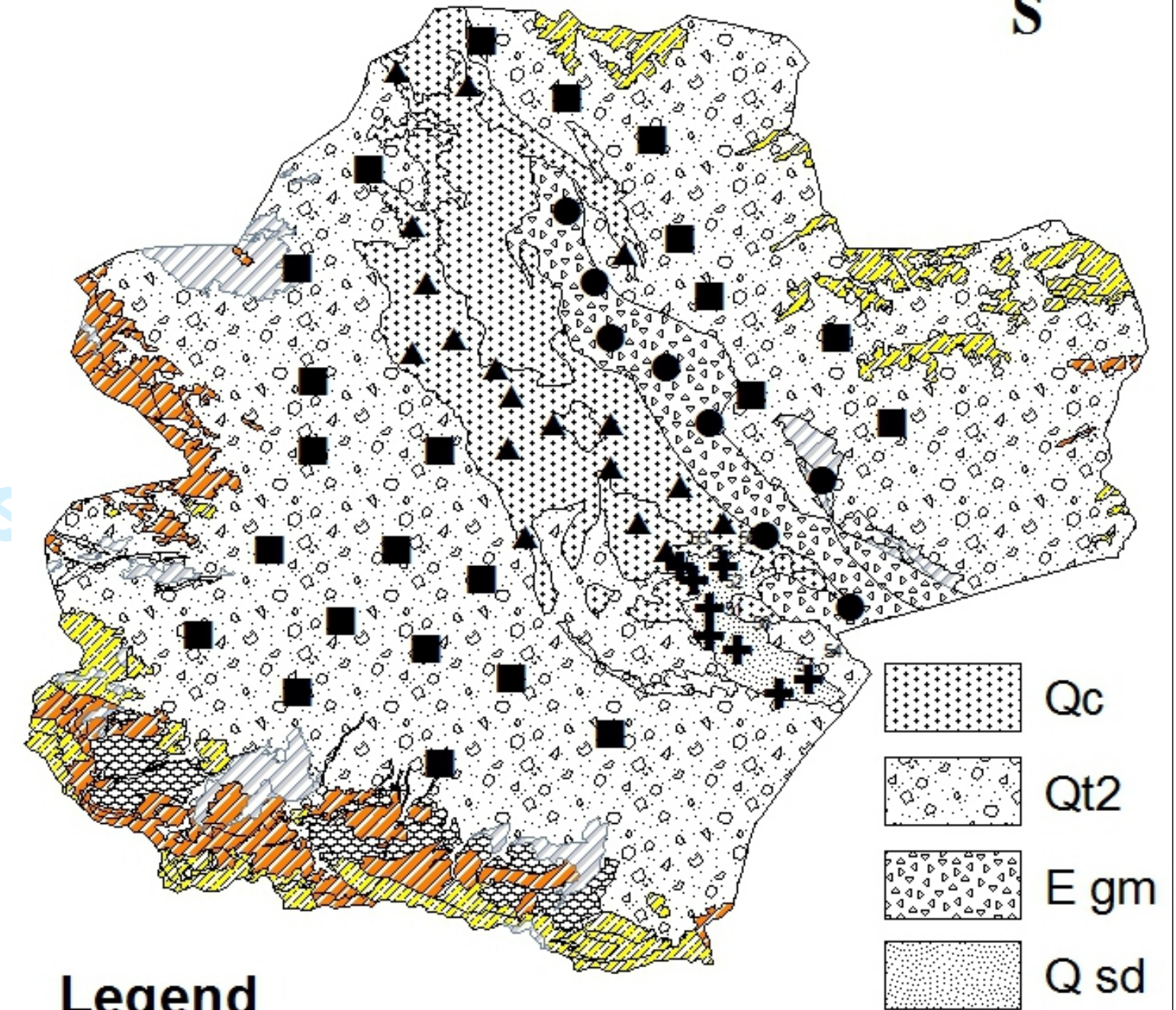
Table 3: Summary geochemistry data for sand dune samples and potential sediment sources. All are reported to two significant figures, except Sr, which is reported to three, due to the larger magnitude of the concentrations.

Table 4: Normality tests on the raw data for the tracers selected for the fingerprint revealed that two tracers (Sr and δCe) were not normally distributed for the Qt2 unit; for this reason, Box-Cox transformations were performed on all data, which did provide normal distributions for all tracers.

Table 5. Estimated contribution from each source for aeolian sediment samples by Bayesian mixing model.



D



Legend

● Egm sample

■ Qt2 sample

▲ Qc sample

+ Sand dune sample

Paleozoic Formations

Cenozoic Formations

Mesozoic Formations

Pre-Cambrian Formations

Qc

Qt2

E gm

Q sd

A. Iran map

B. Yazd Province boundary

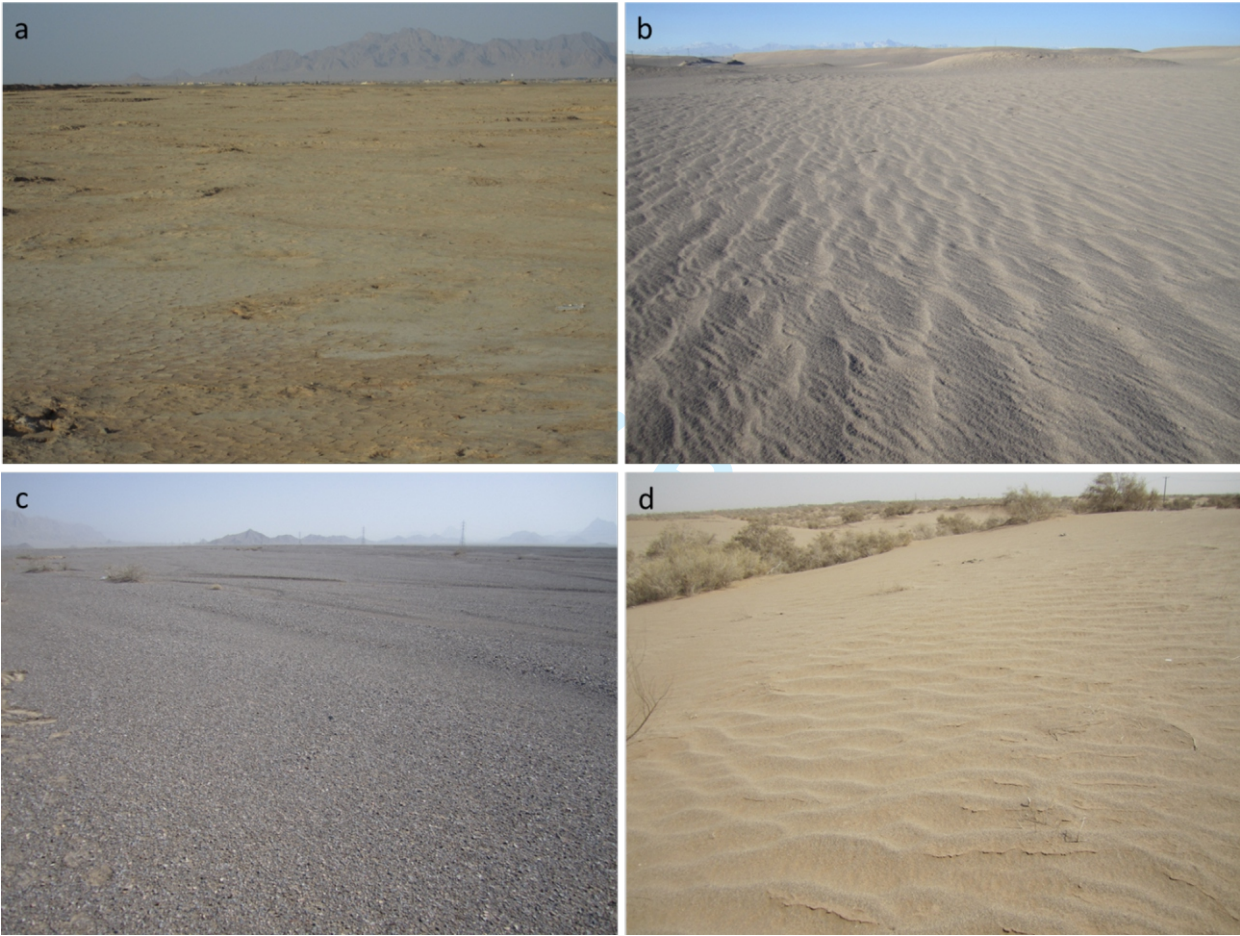
C. Satellite image of Yazd-Ardekan Plain

D. Geological map of Yazd-Ardekan Plain

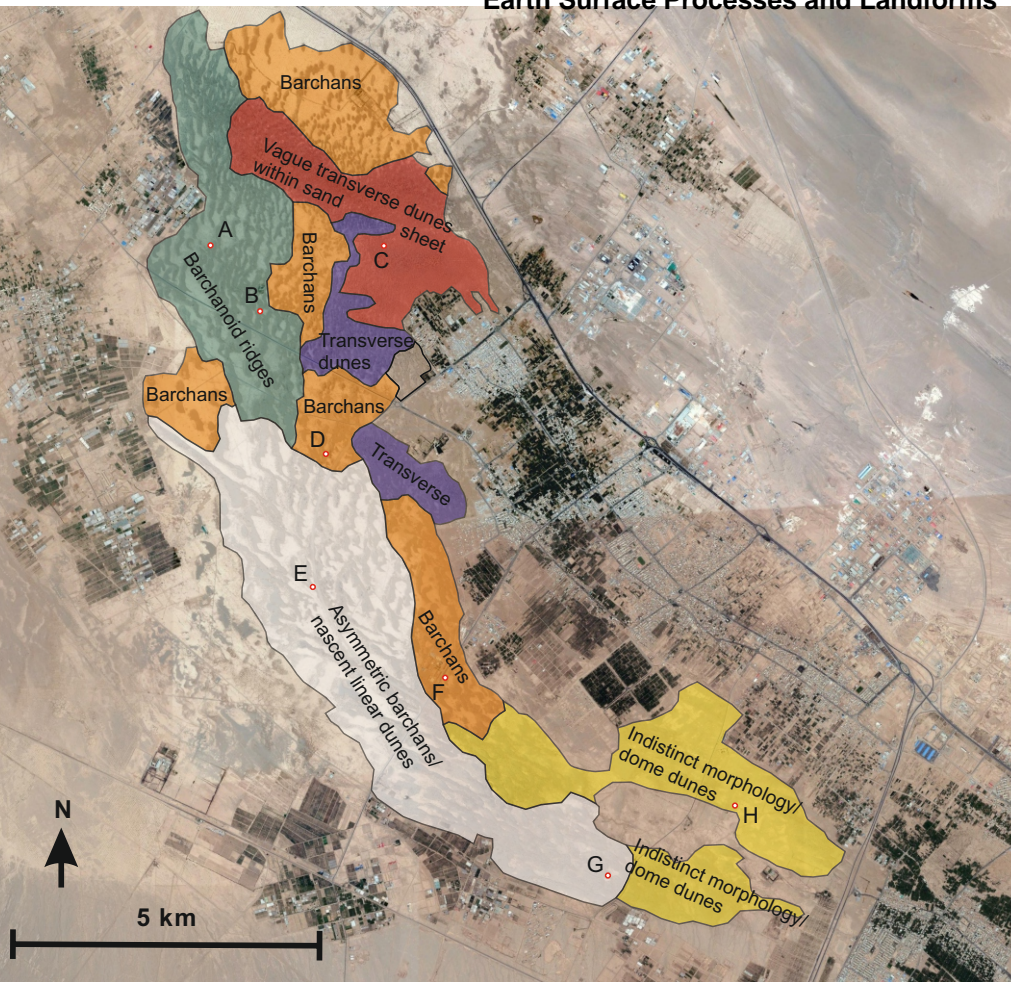
Caspian sea

IRAN

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60

Source

Contribution

Sediment

